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During the three years of ARO support, significant progress has been made toward furthering the understanding of high-intensity laser beam interactions with single liquid droplets with radius, a, much larger than the input laser wavelength (λ_{input}). Our research efforts can be divided into two parts: (1) laser-induced breakdown (LIB) of droplets; and (2) nonlinear optical effects in droplets.

One part of our research was directed toward the optical diagnostics of the plasma associated with the laser-induced breakdown (LIB) processes. We developed several spectroscopic techniques that are capable of providing spatially and temporally resolved measurements of the plasma emission and transmission. The propagation speed and the optical transmission of the breakout plasma from the droplet shadow face and from the illuminated face are determined. The relative heating of the atoms in the plasma is extracted from atomic emission intensities at two different wavelengths.

The other part of our research was directed toward the following nonlinear optical effects: (1) the generation of stimulated Brillouin scattering (SBS) in single droplets; (2) the determination of the angular pattern of stimulated Raman scattering (SRS); (3) the two-photon excitation of lasing droplets: and (4) the modification of the standard nonlinear wave equations to describe the growth and decay of nonlinear waves in droplets.

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Final Report

Richard K. Chang

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CONTENTS

INTRODUCTION

During the three years of ARO support, significant progress has been made toward furthering the understanding of high-intensity laser beam interactions with single liquid droplets with radius, a, much larger than the input laser wavelength (λ_{input}), i.e., droplets with large size parameters $x = 2\pi a/\lambda_{input}$. Our research efforts can be divided into two parts: (1) laser-induced breakdown (LIB) of droplets; and (2) nonlinear optical effects in droplets.

A micrometer-size transparent droplet acts as a thick lens to focus the input radiation just outside the droplet shadow face and to concentrate the input radiation mainly in a region just inside the droplet shadow face. Although the external intensity maximum is an order of magnitude larger than the internal intensity maximum, laser-induced breakdown (LIB) can be initiated within the shadow face of salt-free water droplets with $a < 60$ μm. The addition of salt further lowers the LIB threshold.

One part of our research was directed toward the optical diagnostics of the plasma associated with the LIB processes. We developed several spectroscopic techniques that are capable of providing spatially and temporally resolved measurements of the plasma emission and transmission. The propagation speed and the optical transmission of the breakout plasma from the droplet shadow face and from the illuminated face are determined. The relative heating of the atoms in the plasma is extracted from atomic emission intensities at two different wavelengths.

A micrometer-size transparent droplet also acts as an optical cavity to provide feedback for the internally generated nonlinear radiation. In addition, a droplet with a large size parameter gives rise to a quantum electrodynamic (QED) effect that causes the spontaneous emission coefficient to be enhanced. Consequently, at input laser intensities below the LIB threshold, nonlinear optical process can occur within the droplets. In particular, stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS) can be generated in single droplets because the droplet morphology provides the necessary optical feedback and enhances the spontaneous emission coefficient.

The other part of our research was directed toward the following nonlinear optical effects: (1) the generation of SBS in single droplets; (2) the determination of the angular pattern of SRS from single droplets; (3) the two-photon excitation of lasing droplets: and (4) the modification of the standard nonlinear wave equations to describe the growth and decay of nonlinear waves in droplets. To our knowledge, SBS in droplets was observed for the first time. In addition, the fine structures in the angular pattern of SRS was measured for the first time. The observations of SBS and fine structures in the angular pattern of SRS are important to the overall understanding of nonlinear optical processes in single droplets that are irradiated by a high-intensity laser beam.

RESEARCH ACCOMPLISHMENTS

A brief summary is presented of our main findings related to the LIB process and the generation of SBS and SRS at input intensities below the LIB threshold. Details of our accomplishments can be found in the publications resulting from the research (see page 9).

I. LASER-INDUCED BREAKDOWN (LIB)

(a) Plasma Emission: Temporally and Spatially Resolved Results

We devised a new plasma diagnostic technique which provides both temporally and spatially resolved information at two-selectable wavelengths within the plasma profiles of a single transparent water droplet irradiated by a high-intensity laser beam. The two-selectable wavelengths correspond to the resonance emission of Na and the Balmer H emission or to some emission within the plasma continuum. The measurement of the intensity at two wavelengths as a function of time and space along the z-axis, $I_{\lambda_1}(z,t)$ and

$I_{\lambda_2}(z,t)$, is accomplished by using the following: (1) a spectrograph to disperse the plasma emission; (2) two fiber ribbons, placed at the output plane of the spectrograph, to preserve the spatial integrity of the plasma along the laser propagation direction (z-axis) and to direct the plasma image to the streak camera; and (3) a streak camera to determine the plasma emission intensity as a function of time. The time profile of the input-laser pulse $I_{input}(t)$ is de-termined by directing a portion of the input laser beam on the streak camera.

Preliminary data of the temporally and spatially resolved plasma emission was reported by us in Opt. Lett. 12, 576 (1986). Soon afterwards, J.C. Carls and J.R. Brock published their theoretical findings in Opt. Lett. 13, 273 (1988). Encouraged by the agreement between our experimental findings and their theoretical results, we continued to make additional measurements of the propagation velocity of the plasma plumes along the laser propagation direction at increasing higher input intensities.

The following properties of the laser-induced breakdown plasma are obtained: (1) the propagation speed of the breakout plasma from the droplet shadow and illuminated faces is extracted from the $I_{\lambda_1}(z,t)$ and $I_{\lambda_2}(z,t)$ profiles; (2) the upper limits of the temperature of the atoms in the plasma are deduced from the differences in the $I_{\lambda_1}(z,t)$ and $I_{\lambda_2}(z,t)$ profiles, when λ_1 and λ_2 are set at the emission lines of two different types of atoms, e.g., Na and H which have different excitation potentials; and (3) the shielding of the internal plasma and of the breakout plasma from the illuminated face to prevent further heating of the breakout plasma from the shadow face. The multipulse structure of our multimode Nd:YAG laser turned out to be an additional check of the validity of the one-dimensional electro-hydrodynamic model which has been developed by Prof. James R. Brock. The agreement between the experimental and theoretical findings were reasonably good. Our experimental observations and Prof. Brock's thoretical results appear as two back-to-back papers: J. Opt. Soc. Am. B 8, 319 (1991) and J. Opt. Soc. Am. B 8, 329 (1991).

(b) Plasma Transmission: Temporally and Spatially Resolved Results

A new experimental configuration is devised which enabled us to determine simultaneously the temporal evolution of the plasma transmission T(z,t) from different points along a line in the plasma plume. The near-IR output ($\lambda = 1.064\ \mu m$) of a Nd:YAG laser is used to cause LIB in a single transparent droplet. A time-delayed green-laser radiation (from the green-second harmonic of Nd:YAG) is spit into two parts. One part of the probe beam is sent directly to the upper portion of the streak camera and serves as the reference beam. Another part of the probe beam traverses the droplet and the two plasma plumes, which are ejected from the droplet shadow and illuminated faces, and is then sent to the lower portion of the streak camera. The intensity ratio of the probe beam that traverses the plasma plumes and the reference beam provides information of the plasma attenuation or transmission T(z,t).

From the intensity ratio of the green-probe beam that traversed the plasma and the green-reference beam, we were able to determine the following properties of the expanding plasma: (1) the time development of the one-dimensional resolved transmission of a green beam at different locations along a line parallel to the near-IR laser beam direction, e.g., T(z,t) in front of the droplet shadow face and behind the droplet illuminated face; (2) the propagation speed of the transmission front away from the droplet shadow and illuminated faces; (3) the propagation speed of the fully developed portion of the plasma away from the droplet shadow and illuminated faces; and (4) the decrease of T(z,t) from 100% and its subsequent recovery toward 100%.

II. NONLINEAR OPTICAL EFFECTS

(a) Stimulated Brillouin Scattering

In the case of spontaneous Brillouin scattering from a liquid cell, the Brillouin shift $\Delta\omega$ is dependent on the angle θ between the incident and scattered wave vectors, i.e., $\Delta\omega = \Delta\omega_{max}\sin(\theta/2)$, where $\Delta\omega_{max} = 2\omega_0(v/c)m(\omega)$, where ω_0 is the frequency of the green-incident light, v is the hypersonic acoustic velocity, and $c/m(\omega)$ is the light velocity in the liquid with refractive index $m(\omega)$. When the SBS threshold is exceeded in a liquid cell, an intense acoustic wave is generated and travels toward the laser, i.e., with $\theta = 180°$. Furthermore, the Brillouin-shifted radiation that is generated in a liquid cell without reflectors is also traveling toward the laser with $\theta = 180°$.

The situation is quite different for a liquid droplet. The droplet morphology introduces a spread in the wave vector of the incident beam and hence a spread in θ and $\Delta\omega$. In addition, one or more of the morphology-dependent resonances (MDR's) that exist within the angularly broadened $\Delta\omega$ can provide optical feedback for the internally generated Brillouin radiation. For the Brillouin wave, SBS can only be achieved if the round-trip gain is larger than the round-trip loss.

When the second-harmonic output of a single-mode of a Nd:YAG laser is used to irradiate a single droplet, the SBS spectrum has been spectrally analyzed with a Fabry-Perot interferometer. The intensity thresholds for SBS from micrometer-size water and methanol droplets are found to be remarkably low. In fact, the SBS threshold is 3X lower than the SRS threshold and nearly an order of magnitude lower than the LIB threshold. When intense SBS is generated, the acoustic wave associated with the SBS is not intense enough to shatter the droplets. We were surprised that no SBS was observed for CS_2 droplets, particularly since CS_2 in a cell is known to have the lowest SBS threshold among all liquids.

(b) Angular Scattering Pattern of Stimulated Raman Scattering

A publication from another research laboratory reported that the angular scattering pattern of SRS is smooth, without any lobes. This finding is somewhat disturbing to our overall understanding of SRS, which must be on one or more MDR's in order for the droplet to act as an optical cavity to provide the necessary feedback for the SRS process.

If the SRS radiation is coincident with one or more MDR's, then the angular scattering pattern should be that of the MDR's. In specific, the angular pattern should have 2n peaks as the azimuthal angle is varied from $\phi = 0^\circ$ to 360°, where n = mode number of the MDR.

We have detected many lobes in the angular distribution of SRS $I_{SRS}(\theta)$ from ethanol droplets. In particular, $I_{SRS}(\theta)$ from ethanol droplets is observed to be sinusoidal with n peaks, which are consistent with the mode number n of the MDR's. The existence of fine structures in $I_{SRS}(\theta)$ has two fundamental implications: (1) The two internal counterpropagating SRS waves are coherent, i.e., there is a constant phase difference between the two counterpropagating waves. (2) The internal intensity distribution of the SRS adopts that of the MDR which is providing the feedback for the SRS. We conclude also that the fine structures in $I_{SRS}(\theta)$ can be used to identify the mode number n of the MDR.

(c) Other Nonlinear Optical Effects in Droplets

We have initiated investigation of two other topics of nonlinear optical effects in droplets: (1) two-photon pumped lasing; and (2) nonlinear wave equations appropriate to describe the growth and decay of nonlinear waves in droplets. A brief description of the two unfinished projects is presented.

The two-photon excited fluorescence of dye molecules in droplets is noted to exhibit MDR-related peaks superimposed on the smooth fluorescence emission profile. What is significant is that we observed fluorescence emission in the blue when a red-laser beam is used to pump the dye absorption band in the deep blue. This motivated us to attempt to investigate the possibility of two-photon pumped lasing in single droplets. Lasing emission has been observed with two-photon excitation of several different dyes dissolved in ethanol and methanol. Standard 35 mm photographs show a red input beam incident on the droplet and blue radiation emerging from the rim of the droplet. Using a multimode Q-switched Nd:YAG laser, we investigated the dye concentration and threshold dependence on the input-laser intensity. However, because of the multimode nature of the pumped laser, the resultant dye laser output intensity exhibited a significant amount of fluctuation. In the future, we plan to use a single-mode Q-switched Nd:YAG laser as the pump laser and remeasure the dependence of the two-photon pumped dye-laser intensity on the dye concentration and the input-laser intensity.

The standard equations for the growth and coupling of nonlinear waves in a liquid cell are one dimensional and are in the direction of the input and nonlinear beams (along the z axis). For a droplet, the input-pump intensity is localized in a small region along the principal diameter (defined as the droplet z axis) just within the droplet shadow face, and the resultant nonlinear waves propagate around the rim. The nonlinear wave resonant with one MDR is a standing wave, which can be decomposed into two counterpropagating waves traveling around the droplet rim. Thus, for the growth and coupling of the nonlinear process in a droplet, conventional one-dimensional wave equations in an optical cell must be modified as one-dimensional wave equations with the waves propagating around the droplet rim (i.e., a function of θ and ϕ).

In the computer simulations, we included the growth of stimulated Brillouin scattering (SBS) or stimulated Raman scattering (SRS) starting from the spontaneous noise and from the parametric noise generated by the four-wave mixing process. The time dependence of the observed input-laser pulse is simulated in our calculation and the radial dependence of the input laser intensity is determined by the Lorenz-Mie theory. For simplicity, we assumed that the laser is undepleted as a result of pumping the nonlinear waves. For each complete trip around the droplet circumference, the Brillouin or Raman wave experiences gain as it traverses the laser pump region. In addition, during each round trip, the Brillouin or Raman wave experiences losses associated with linear absorption, leakage commensurate with the Q of the MDR, and depletion caused by coupling to other nonlinear waves.

Remarkable agreement between our preliminary computer simluation and the experimentally observed results has been achieved for the following: (1) the time delay between the start of the input laser pulse and the SBS or SRS pulse; (2) the time delay between the first-order Stokes SRS pulse and the (n)th-order SRS pulses; (3) the correlated growth and decay of the SBS and the first-order Stokes SRS pulses; (3) the correlated growth of the first-order Stokes SRS with the second-order Stokes SRS as well as between the second-order Stokes SRS and the third-order Stokes SRS. We conclude that the modifications of the nonlinear wave equations, which were originally developed for the liquid cell, can account for the nonlinear processes in a single liquid droplet. We plan to submit a manuscript describing our modified wave equations and computer simulations on the time profiles of the SBS and SRS pulses.

PUBLICATIONS RESULTING FROM THE RESEARCH

Laser-Induced Breakdown

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Nonlinear Optical Effects

5. J.-Z. Zhang and R.K. Chang, "Generation and Suppression of Stimulated Brillouin Scattering in Single Liquid Droplets," J. Opt. Soc. Am. B 6, 151 (1989).

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